PHYSICAL REVIEW B, VOLUME 65, 161101(R)

Optical conductivity of the heavy fermion superconductor $CeCoIn_5$

E. J. Singley, D. N. Basov, E. D. Bauer, and M. B. Maple
Department of Physics, University of California–San Diego, La Jolla, California 92093-0319

(Received 13 December 2001; published 5 April 2002)

The electrodynamic response of the heavy fermion compound $CeCoIn_5$ is studied via infrared spectroscopy. Below 100 K the high temperature Drude-like conductivity evolves into two distinct components. At low energies a narrow mode is observed which is characteristic of the response of heavy quasiparticles. At higher energies a gap structure emerges with $2\Delta\sim600\ \mathrm{cm}^{-1}$ (75 meV). Examination of the fine features of the optical conductivity reveals additional absorption below the gap. This intergap resonance can be qualitatively understood as a Holstein band, where the quasiparticles are strongly coupled to a collective mode at $\sim65\ \mathrm{cm}^{-1}$ (8 meV).

DOI: 10.1103/PhysRevB.65.161101 PACS number(s): 71.27.+a, 74.70.Tx, 78.20.Ci

Recently a new class of heavy fermion compounds with chemical formula $CeMIn_5$ has been discovered, where M can be either Rh, Ir, or Co.[1–3] The ground state of these materials depends strongly on the transition metal M, being antiferromagnetic for Rh but superconducting for Ir ($T_c=0.4$ K) and Co ($T_c=2.3$ K). Alloying the different members of this family produces a rich phase diagram of superconducting and antiferromagnetic regions that can coexist with each other.[4,5] One way to view these materials is as a composite of alternating $CeIn_3$ and MIn_2 layers, where $CeIn_3$ is itself a three-dimensional heavy fermion superconductor ($T_c=200$ mK at 25 kbar[6]). In this fashion these materials may be similar to cuprate superconductors where reduced dimensionality and the proximity to an antiferromagnetic phase are believed to contribute to the enhanced values of T_c.[7]

Like the cuprates, superconductivity in $CeMIn_5$ compounds is believed to be unconventional. Specific heat,[2] nuclear magnetic resonance (NMR),[8] and thermal conductivity[9] experiments are all consistent with an order parameter with lines of nodes. Additionally, observation of a fourfold symmetry of the thermal conductivity in a rotating magnetic field indicates a $d_{x^2-y^2}$ order parameter in $CeCoIn_5$.[10] A question which is still open is the nature of the pairing interaction. The close association between antiferromagnetic order and superconductivity in heavy fermion systems suggests that magnetic excitations may play an important role in pairing the electrons.[11] In fact, tunneling experiments on UPd_2Al_3 have found strong coupling effects on the conductance spectrum in an energy region where magnetic excitations have also been observed by neutron scattering.[12] While the spin fluctuation spectrum is as yet unknown in $CeCoIn_5$, the optical conductivity can act as an indirect probe of these excitations,[13–15] in an analogous manner to tunneling spectroscopy. An additional consideration in the study of heavy fermion superconductors, is that the low temperature ordered phase is formed out of a coherent many-body state.[16] The analysis of the optical conductivity is an ideal tool for unfolding the energy scales associated with this collective regime.[17,18]

In this Rapid Communication, we present the results of an infrared study of $CeCoIn_5$ at $T>T_c$. An analysis of the frequency dependent conductivity, $\sigma_1(\omega)$, reveals the opening of a hybridization gap at $T\sim100$ K, with magnitude $2\Delta\sim600\ \mathrm{cm}^{-1}$ (75 meV). In the very far infrared, we also observe a mode characteristic of the response of heavy quasiparticles, from which we deduce a mass enhancement of $m^*=26–55m_b$, where m_b is the band mass of the charge carriers. Further analysis of $\sigma_1(\omega)$ in the coherent state shows the possible signature of a Holstein band. We suggest that this feature may be due to quasiparticles interacting with antiferromagnetic fluctuations with an energy scale of $\sim65\ \mathrm{cm}^{-1}$ (8 meV).

Single crystals of $CeCoIn_5$ were grown using an Indium flux method.[4] The crystals were approximately 5 mm^2 in the ab-plane orientation and 50μm thick along the c axis. The as-grown crystals had smooth, shiny surfaces, and no polishing was required for the measurement. The near normal reflectance of the ab-plane was measured from the far-infrared (FIR) to the near-ultraviolet. A Fourier transform spectrometer was used from 30 cm^{-1} to 18,000 cm^{-1}, and a grating monochromator was used from 12,000 cm^{-1} to 50,000 cm^{-1}. The complex optical constants were obtained from the measured reflectance using the Kramers-Kronig relations. Details of the experimental procedure can be found in Ref. 19.

The inset of Fig. 1 shows the reflectance, $R(\omega)$, up to 50,000 cm^{-1} on a logarithmic scale. In nearly the entire spectral range $R(\omega)$ is above 50%. Two distinct contributions to the reflectance can be resolved in the data. Below $\omega\simeq10{,}000\ \mathrm{cm}^{-1}$, $R(\omega)$ is characterized by a metallic response where $R(\omega)\rightarrow1$ as $\omega\rightarrow0$. At $\omega\simeq17{,}000\ \mathrm{cm}^{-1}$ a peak is seen in $R(\omega)$ which is indicative of interband transitions. The main panel of Fig. 1 shows the temperature dependence of $R(\omega)$ below 2,000 cm^{-1}. At 292 K [thin black line] $R(\omega)$ increases monotonically with decreasing frequency. As the temperature decreases to 100 K, a minimum begins to develop near 500 cm^{-1}. This minimum deepens as the temperature is reduced further.

The temperature dependent dc conductivity, $\sigma_1(\omega=0)$, normalized to the value at 300 K is shown in the inset of Fig. 2 on a log-log plot.[20] As the temperature decreases the dc conductivity increases slightly at 200 K, then decreases back to the high temperature value at 50 K. Below 50 K the dc conductivity rises dramatically, increasing by nearly an order of magnitude before the onset of the superconducting transition at 2.3 K. This sharp rise in the dc conductivity is a

©2002 The American Physical Society

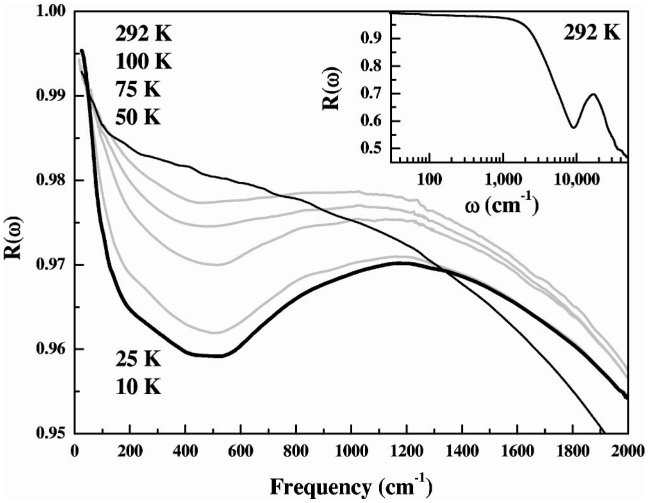

FIG. 1. Temperature dependence of the reflectance in the far infrared. At 292 K, $R(\omega)$ increases smoothly showing metallic behavior. At lower temperatures a minimum develops in $R(\omega)$ at 500 cm^{-1}. The inset shows the reflectance at 292 K over the entire measured frequency range on a log scale.

hallmark of the formation of a coherent many-body state in heavy fermion systems. The source of this coherence lies in the Kondo interaction which causes a hybridization of carries in the conduction band with localized *f*-electrons. This hybridization process leads to a reduction in scattering for carriers near the Fermi energy, and therefore an increase in the dc conductivity. Additional consequences of this interaction, is an increase in the quasiparticles mass and the formation of a gap in the density of states (DOS).[16]

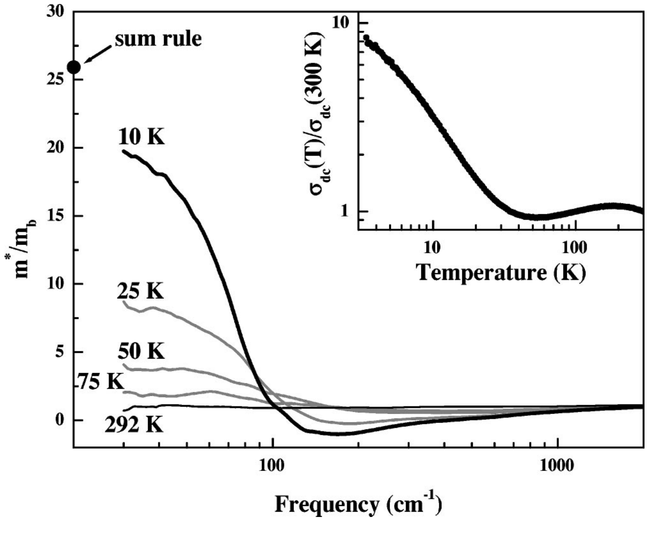

FIG. 2. Inset: dc conductivity normalized to the high temperature value on a log-log scale. Main panel: Frequency dependence of the effective mass on a log frequency scale. At 292 K, $m^*\simeq m_b$ over the entire spectral range. At lower temperatures the effective mass is also equal to the band mass at high energies. Below 100 cm^{-1}, m^* increases rapidly with decreasing temperature and frequency. The filled circle represents the value of m^*/m_b at 10 K obtained from Eq. 1.

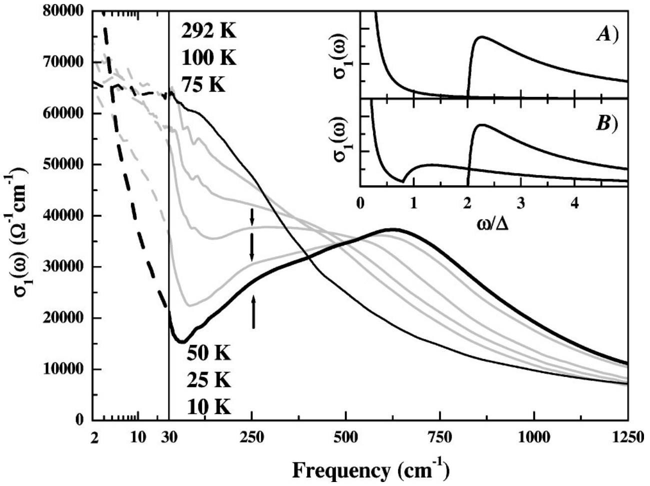

FIG. 3. Dissipative part of the conductivity in the far infrared. The low frequency part of the graph shows the conductivity in the region corresponding to the extrapolated reflectance on a log scale. At 292 K, $\sigma_1(\omega)$ shows a metallic Drude-like response. At low temperatures the development of a gap in the DOS is apparent from the peak in $\sigma_1(\omega)$ which grows near 600 cm^{-1}. The very narrow mode at low energies is a signature of the intraband response of the heavy quasiparticles. The arrows near 250 cm^{-1} highlight additional absorption within the gap. Panel (A) of the inset shows model calculations of $\sigma_1(\omega)$ for a hybridization gap and heavy quasiparticle intraband response (Ref. 25). In panel (B) the heavy quasiparticle component is allowed to couple to a boson mode at $\omega_B=0.75\Delta$ which produces an additional absorption band.

The evolution of the electronic states as the sample enters the coherent state is more clearly revealed with the analysis of the frequency dependent conductivity, shown in the main panel of Fig. 3. At 292 K, $\sigma_1(\omega)$ shows canonical metallic behavior, with a line shape in accord with Drude theory. As the temperature is lowered to 100 K strong deviations in the spectrum of $\sigma_1(\omega)$ become evident. At frequencies below $\simeq 300$ cm^{-1} $\sigma_1(\omega)$ is reduced from the high temperature value, while at higher energies an increase in $\sigma_1(\omega)$ is observed. This trend in the deconstruction of the Drude spectrum continues to grow as the temperature is further reduced. By 10 K (thick black line) two distinct components can be resolved in the frequency dependence of the conductivity which clearly exhibit the key features of the coherent many body state. At the lowest measured energies, the tail of a narrow peak can be seen, which represents the intraband response of the heavy quasiparticles. The second component is the peak in $\sigma_1(\omega)$ centered near 600 cm^{-1}, which signals electronic absorption across a gap in the DOS. Both of these features reflect the radical changes which occur in the electronic system as the conduction band carriers hybridize with the localized *f*-electrons.

The low frequency region of the main panel of Fig. 3 shows $\sigma_1(\omega)$ corresponding to the extrapolated region of the reflectance on a logarithmic scale. While different low energy extrapolations of $R(\omega)$ will slightly alter the behavior of $\sigma_1(\omega)$ below 30 cm^{-1}, the qualitative result remains unchanged: a very narrow mode exists at 10 K which crosses the 292 K conductivity curve, and is roughly a factor of two greater in magnitude at 2 cm^{-1}, in accord with the dc transport data.

An analysis of the low energy mode reveals the mass enhancement of the quasiparticles in the coherent state. This mass enhancement may be estimated by applying a sum rule to the $\sigma_1(\omega)$ data. Specifically, the area confined under the conductivity curve is proportional to the density of carriers divided by their effective mass. Therefore assuming the density of carriers remains constant and using the fact that $m^*(292\ \mathrm{K})=m_b$, one obtains:

$$m^*/m_b=\frac{\int_0^{\omega_c^{292\ \mathrm{K}}}\sigma_1(\omega,292\ \mathrm{K})d\omega}{\int_0^{\omega_c^{10\ \mathrm{K}}}\sigma_1(\omega,10\ \mathrm{K})d\omega}, \qquad (1)$$

where $\omega_c^{10\ \mathrm{K}}$ and $\omega_c^{292\ \mathrm{K}}$ are the integration limits at the respective temperatures. These integration limits are chosen such that only the intraband component of $\sigma_1(\omega)$ is included at each temperature ($65\ \mathrm{cm}^{-1}$ at 10 K and $2500\ \mathrm{cm}^{-1}$ at 292 K). Using this procedure a value of $m^*=26m_b$ is obtained for the effective mass at 10 K. An alternative method of estimating the mass enhancement may be derived from the frequency dependence of the complex conductivity, which yields a frequency dependent effective mass.[17,18] The main panel of Fig. 2 shows $m^*(\omega)$ normalized to the band mass at several temperatures. At high energies ($2000\ \mathrm{cm}^{-1}$), the effective mass is just the band mass for all temperatures. The spectrum at 292 K remains essentially constant throughout the displayed frequency range. In contrast, at temperatures where σ_{dc} and $\sigma_1(\omega)$ show signatures of the coherent state, $m^*(\omega)$ increases rapidly below $100\ \mathrm{cm}^{-1}$. At 10 K and the lowest measured frequency $m^*/m_b=20$, while the zero frequency limit yields a value of $m^*/m_b=40$–55, depending on the form of the low frequency extrapolated reflectance used in the Kramers-Kronig analysis.

The values we obtain for the enhanced mass are in reasonable agreement with specific heat results[2] which find $\gamma=290\ \mathrm{mJ/mol\,K}^2$ and de Haas–van Alphen (dHvA) measurements[22] which obtain values of m^* between 5 and 87. It should be noted that an exact comparison of our results to the thermodynamic mass enhancement and the dHvA results is only appropriate for our data in the limit $\omega\rightarrow 0$. Therefore the $30\ \mathrm{cm}^{-1}$ value of $m^*=20$ is almost certainly an underestimate while the $\omega=0$ value suffers due to uncertainties in the extrapolated region of the spectrum.

We now turn to a discussion of the second main feature in the low temperature spectrum of Fig. 3: the absorption maximum observed near $600\ \mathrm{cm}^{-1}$. A key feature in the data is the evolution of spectral weight [area under $\sigma_1(\omega)$ curve] with decreasing temperature. Below $\sim 400\ \mathrm{cm}^{-1}$ the spectral weight is reduced with decreasing temperature while concomitantly increasing in the energy region near $600\ \mathrm{cm}^{-1}$. This behavior signals the opening of a gap in the DOS. While the $600\ \mathrm{cm}^{-1}$ peak observed at 10 K does soften as the temperature increases, it is unclear whether this is because of a temperature dependent gap, or rather just an effect of the spectral weight transfer discussed above. In other related systems the hybridization gap has been found to be independent of temperature.[23,24] The frequency and temperature dependence of $\sigma_1(\omega)$ observed here is commonly found in the low temperature spectrum of heavy fermion materials.[17,18] The gap in the low temperature DOS is a consequence of the hybridization between localized *f*-electrons and the conduction band carriers.[16] What is uncommon about the low temperature spectra in Fig. 3 is the frequency dependence of $\sigma_1(\omega)$ below the $600\ \mathrm{cm}^{-1}$ peak. Examination of the curves reveals additional absorption. Looking at the 10 K spectrum where the hybridization gap is well developed, a shoulder can be seen near $250\ \mathrm{cm}^{-1}$ (marked with arrow). The $250\ \mathrm{cm}^{-1}$ structure is also clearly visible at 25 K and 50 K, and less obviously at 75 K. The position of the shoulder remains essentially unchanged with increasing temperature. Qualitatively, a sharp drop in $\sigma_1(\omega)$ is expected below the hybridization gap.[18,21] Panel (A) of the inset in Fig. 3 illustrates the expected frequency dependence of the low temperature spectrum of $\sigma_1(\omega)$.[25] The two components corresponding to the intraband response of the heavy quasiparticles and the hybridization gap are often adequate in describing $\sigma_1(\omega)$ of heavy fermion materials in the coherent state.[18] However these two components alone cannot account for the frequency dependence of the low temperature data in Fig. 3. The slow decrease of the experimental data below the hybridization peak, along with the shoulder at $250\ \mathrm{cm}^{-1}$ indicates that there is an additional absorption channel below the hybridization gap.

Some insight into the origin of this intergap absorption may be found in specific heat experiments. Measurements of $\Delta C/\gamma T_c$ indicate that CeCoIn$_5$ is a strongly coupled superconductor.[2,4] Indeed, values of $\Delta C/\gamma T_c$ as high as 4.9 have been reported,[4] whereas in the BCS weak coupling limit a value of 1.43 is expected. Independent of the origin of the boson mode which couples the electrons to produce superconductivity, a characteristic absorption feature, known as a Holstein band, should be observed in the optical conductivity above the energy of the mode. This is illustrated in Panel (B) of the inset of Fig. 3 where the quasiparticles that give rise to the narrow low energy mode in Panel (A), are now allowed to couple to a boson mode at $\omega_B=0.75\Delta$. Above ω_B an additional scattering channel is available giving rise to an absorption feature in $\sigma_1(\omega)$. Note that the position of the boson mode is not given by the absorption maximum in $\sigma_1(\omega)$, but rather the minimum between the zero energy mode and intergap resonance. At least qualitatively, the different components of $\sigma_1(\omega)$ shown in Panel (B) can account for the main features of the data observed in Fig. 3.[26] From the spectra of $\sigma_1(\omega)$ in Fig. 3 we estimate the energy of the boson mode to be near $65\ \mathrm{cm}^{-1}$ (8 meV).[27]

A common source of Holstein bands in metals is the electron phonon interaction. While phonons are obvious candidates for the boson mode producing the Holstein band, knowledge of the phonon density of states is required before a direct connection can be made. An alternative origin of the boson mode which strongly couples to the charge carriers is antiferromagnetic fluctuations. While these fluctuations have yet to be directly observed in CeCoIn$_5$, such a hypothesis is worth exploring for several reasons. First of all, the phase diagram of CeMIn$_5$ displays a close proximity between antiferromagnetic order and superconductivity. In fact in the compound $CeCo_{0.5}Rh_{0.5}In_5$ both superconductivity and antiferromagnetism coexist (likewise in $CeIr_{0.5}Rh_{0.5}In_5$).[4,5] Additionally, strong coupling effects attributable to antiferromagnetic fluctuations have already been observed in the superconducting heavy fermion compound UPd_2Al_3.[12] In $CeCoIn_5$, the results of thermal conductivity measurements are also in accord with strong coupling to antiferromagnetic fluctuations.[9,10] It is valuable to once again return to the analogy with the cuprates where there is compelling evidence for a tie between superconductivity and antiferromagnetism.[28] In nearly all families of cuprate superconductors a feature that can be interpreted in terms of a Holstein band is observed in the far infrared spectrum of $\sigma_1(\omega)$.[29] In one of these families ($YBa_2Cu_3O_{7-\delta}$) a quantitative match has been made between the spectrum of the antiferromagnetic fluctuations derived from the Holstein band in $\sigma_1(\omega)$ and direct measurements obtained by neutron scattering.[14] While the close proximity to the hybridization gap does not allow us to determine the spectrum of the collective mode, the energy position of this feature suggest that direct magnetic probes, such as neutron scattering, should focus on the energy range near 65 cm^{-1} (8 meV) when searching for antiferromagnetic fluctuations. Finally we note that in this scenario the disappearance of the Holstein band above the coherence temperature may in part be explained by an increase of spin damping when the hybridization gap is suppressed.[30]

In summary we have studied the electromagnetic response of the heavy fermion compound $CeCoIn_5$. We observe clear signatures of the hybridization state; a gap with $2\Delta \sim 600$ cm^{-1}, and a low energy mode due to the intraband response of the heavy quasiparticles from which we estimate the effective mass to be 26–55m_b. A close inspection of the spectrum of $\sigma_1(\omega)$ reveals additional absorption which may be a result of charge carriers coupling to a collective mode. We estimate the energy of the collective mode to be $\omega_B \simeq 65$ cm^{-1} (8 meV) and suggest that its origin may be antiferromagnetic fluctuations.

The authors would like to thank Sasa Dordevic for useful discussions. This work was supported by NSF and the DOE and the Alfred P. Sloan Foundation. D. N. Basov is a Cotrell Fellow of the Research Corporation.

---

[1] H. Hegger *et al.*, Phys. Rev. Lett. **84**, 4986 (2001).

[2] C. Petrovic *et al.*, J. Phys.: Condens. Matter **13**, L337 (2001).

[3] C. Petrovic *et al.*, Europhys. Lett. **53**, 354 (2001).

[4] V. S. Zapf *et al.*, Phys. Rev. B **65**, 014506 (2002).

[5] P.G. Pagliuso *et al.*, cond-mat/0107266 (unpublished).

[6] N.D. Mathur *et al.*, Nature (London) **394**, 39 (1998).

[7] P. Monthoux and D. Pines, Phys. Rev. Lett. **69**, 961 (1992); P. Monthoux and G.G. Lonzarich, Phys. Rev. B **63**, 054529 (2001).

[8] Y. Kohori *et al.*, Phys. Rev. B **64**, 134526 (2001).

[9] R. Movshovich *et al.*, Phys. Rev. Lett. **86**, 5152 (2001).

[10] K. Izawa *et al.*, Phys. Rev. Lett. **87**, 057002 (2001).

[11] D.L. Cox and M.B. Maple, Phys. Today **48** (2), 32 (1995).

[12] M. Jourdan, M. Huth, and H. Adrian, Nature (London) **398**, 47 (1999).

[13] B. Farnworth and T. Timusk, Phys. Rev. B **14**, 5119 (1976).

[14] J.P. Carbotte, E. Schachinger, and D.N. Basov, Nature (London) **401**, 354 (1999); E. Schachinger, J.P. Carbotte, and D.N. Basov, Europhys. Lett. **54**, 380 (2001); A. Abanov, A.V. Chubukov, and J. Schmalian, Phys. Rev. B **63**, 180510 (2001).

[15] M.G. Hildebrand *et al.*, Phys. Rev. Lett. **87**, 227002 (2001).

[16] For a review see: A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, 1997); P. Fulde, *Electron Correlations in Molecules and Solids* (Springer-Verlag, Berlin, 1993); N. Grewe and F. Steglich, in *Handbook on the Physics and Chemistry of Rare Earths*, Vol. 14, edited by K. A. Gschneidner and L. Eyring (Elsevier Science, Amsterdam, 1994).

[17] L. Degiorgi, Rev. Mod. Phys. **71**, 687 (1999).

[18] S.V. Dordevic *et al.*, Phys. Rev. Lett. **86**, 684 (2001).

[19] C.C. Homes *et al.*, Appl. Opt. **32**, 2976 (1993).

[20] The dc conductivity was measured using a four probe technique on the ab-face of several crystals grown in the same batch as the one used for reflectance measurements. All samples had a similar temperature dependence, whereas variations in the absolute value were within 20%.

[21] D. Hall *et al.*, Phys. Rev. B **64**, 212508 (2001); R. Settai *et al.* (unpublished).

[22] Z. Schlesinger *et al.*, Phys. Rev. Lett. **71**, 1748 (1993).

[23] B. Bucher *et al.*, Phys. Rev. Lett. **72**, 522 (1994).

[24] P. Coleman, Phys. Rev. Lett. **59**, 1026 (1987).

[25] The zero energy absorption peaks are Lorentzian in accord with Drude theory. The hybridization gap was calculated using the BCS formalism with type 1 coherence factors. See, for example, M. Tinkham, *Introduction To Superconductivity* (McGraw-Hill, New York, 1996). The Holstein band was calculated using a delta function as the boson spectrum for simplicity. For details of the calculation see A. V. Puchkov, D. N. Basov, and T. Timusk, J. Phys: Condens. Matter **8**, 10 049 (1996).

[26] Recent NMR experiments have observed crystal field effects in $CeCoIn_5$ [N.J. Curro *et al.*, Phys. Rev. B **64**, 180514 (2001)]. The energy splitting of these levels was found to be 34 K (~ 25 cm^{-1}) and 102 K (~ 70 cm^{-1}). While crystal field effects may play an important role in $CeCoIn_5$, the magnitude of the splitting as observed by NMR indicates that they are not likely to be responsible for the 250 cm^{-1}, feature in $\sigma_1(\omega)$.

[27] This may be somewhat of an overestimate, since at 10 K the formation of the hybridization gap is incomplete. As $T \rightarrow 0$, additional low energy spectral weight will be transferred above 600 cm^{-1}, resulting in a shift of the minimum to lower frequencies. This trend can be observed as the temperature is lowered from 50 K to 10 K.

[28] P. Dai *et al.*, Nature (London) **406**, 965 (2000); M. Eschrig and M. R. Norman, Phys. Rev. Lett. **85**, 3261 (2000).

[29] E.J. Singley *et al.*, Phys. Rev. B **64**, 224503 (2001).

[30] D.K. Morr and D. Pines, Phys. Rev. Lett. **81**, 1086 (1998).